Fluid Dynamics &
Materials Processing

Tech Science Press

DOI: 10.32604/fdmp.2021.016265

ARTICLE

Analysis of the Thermal Behavior of a Lithium Cell Undergoing Thermal Runaway

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Received: 21 February 2021 Accepted: 13 April 2021

ABSTRACT

This study examines the thermal runaway of a lithium ion battery caused by poor heat dissipation performances. The heat transfer process is analyzed on the basis of standard theoretical concepts. Water mist additives are considered as a tool to suppress the thermal runaway process. The ensuing behaviour of the battery in terms of surface temperature and heat generation is analyzed for different charge and discharge rates. It is found that when the remaining charge is 100%, the heat generation rate of the battery is the lowest, and the surface temperature with a 2C charge rate is higher than that obtained for a 0.5C charge rate. The experimental results show that when the additive concentration is 20% NaCl, its ability to inhibit the thermal runaway is the strongest.

KEYWORDS

Lithium ion battery; thermal runaway; discharge rate; heat generating rate; water mist

1 Introduction

In recent years, people's demand for industrial production and energy has grown ever larger with the rapid development of the global economy and science and technology. People's unrestrained exploitation and use of non-renewable resources make the energy situation tenser and tenser. In order to alleviate energy tension, the development of renewable resources has become a global research hotspot, and the battery has gradually become a necessity of production and life. The main problem in battery development is its service life, and the thermal runaway phenomenon o is the biggest factor affecting this service life. Hence, it is of great importance for future sustainable battery development to restrain the thermal runaway phenomenon.

The lithium ion battery is currently recognized as a form of recyclable clean energy which has great development potential for the future. In order to understand the status of research into the lithium ion battery, this paper reviews recent relevant research. Tab. 1 is a summary of recent research into improvement of the lithium ion battery, which provides a basis for follow-up research.

The results from Tab. 1 shown that the energy transfer process of a lithium ion battery after thermal runaway will damage the performance of the battery and affect its service life. Controlling and preventing such thermal runaway is the focus of future research. In order to control the thermal runaway phenomenon in lithium ion batteries, based on the heat transfer theory, the energy transfer form of the lithium ion battery after thermal runaway is studied in this paper. Water mist additives are used to control and improve the thermal runaway phenomenon, enhance the batteries' service life, improve their performance, and enhance their development prospects in the future.

Table 1: Research progress on improvement of lithium ion battery

Researchers	Time	Research contents
Ning et al. [1]	2020	A simulation method for thermal runaway behavior of lithium ion batteries was provided, through which the surface temperature change and the thermal reaction of electrode material could be obtained. The thermal runaway temperature and thermal runaway mechanism of the battery were analyzed through the model and fitting parameters were obtained.
Maheswari et al. [2]	2020	A unique thermal control strategy to improve aging of the battery was proposed, mainly to control the internal temperature of the battery used in electric vehicles (EV) within the range of 20°C–40°C, and reduce the frequent voltage fluctuation in the DC bus, so as to increase the life of the storage battery. The unique thermal control strategy of the hybrid EV system can effectively control the frequent charge and discharge of the lithium ion battery, and eliminate thermal runaway problems such as leakage, smoke, exhaust, rapid disassembly and flames.
Bugryniec et al. [3]	2020	A computational method was proposed to investigate potential thermal runaway propagation (TRP) in LFP cathode batteries. It was mainly based on the 2D model of the battery pack. If one of the batteries experiences an internal short circuit, thermal runaway propagation will not occur even under various extreme environmental conditions, which indicates that controlling the battery with LFP as cathode can realize the potential of battery safety and large-scale use.
Zhang et al. [4]	2020	A composite barrier technology was proposed, and the soft clad nickel cobalt manganese (NCM811) battery was taken as the main experimental research object to determine the relationship between the surface temperature of the battery and thermal runaway, as well as the relationship between microstructure and thermal barrier properties. The study shows that the thermal barrier effect is greatest in the thermal barrier test of microstructure-size materials with a millimeter thickness.

2 Method

2.1 Structure and Mechanism of Lithium Ion Battery

According to the arrangement of internal materials, lithium ion batteries can be divided into cylindrical and square structures. The most commonly used is the square battery, which is stacked by the chemical reaction mechanism of the battery [5]. A lithium ion battery is generally composed of positive and negative electrodes, positive and negative materials, electrolyte solution, a separator and other parts.

Positive and negative electrodes: Generally, aluminum foil and copper foil are used as the current collectors for positive and negative electrodes to connect with the external circuit [6].

Positive and negative materials: Positive materials are the main supplier of $Li+$ in lithium ion batteries. Lithium transition metal oxides are the positive materials studied and used most, such as lithium cobaltate, lithium manganate, LFP, and ternary. Lithium metal was used as the negative material in the initial research of these batteries. However, the specific capacity of lithium metal is too high when it is used as the negative electrode; moreover, it is easy to generate dendrite when the battery is charged, resulting in a short circuit of the battery. Therefore, nowadays, graphite, carbon and metal oxide are used as the negative electrode material of the battery, instead [7].

Electrolyte solution: The principal types of electrolyte in lithium ion batteries are liquid electrolytes and solid electrolytes. The more commonly used electrolyte is the liquid form, which means an electrolyte solution, an important transfer station for the chemical reaction in a lithium ion battery. It is responsible for transferring the charge between positive and negative electrodes. The composition, structure and chemical ratio of the electrolyte determine the performance of the electrolyte, which ultimately determines the performance of the lithium ion battery.

Fig. 1 presents the specific structure. In this structure, the main function of the positive and negative electrodes is to provide anions and cations. In lithium ion batteries, the capacity of the battery is often determined by the capacity for lithium ion removal and insertion in the negative materials [8,9].

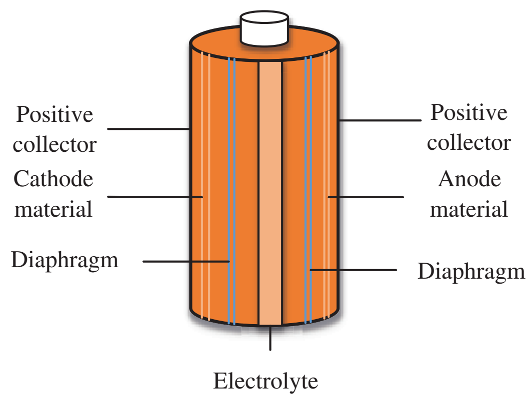

Figure 1: Specific structure of lithium ion battery

In the discharging process of the lithium ion battery, the anions on the cathode material come out and embed into the surface of the cathode material. The LEP battery is taken as an example. During the charging process of the battery, Li^+ migrates from the $[FePO_4]^-$ layer of the cathode material, passes through the electrolyte layer of the battery and enters the negative electrode material of the battery. Fe^{2+} on the negative electrode material is oxidized to Fe^{3+}, and the electrons reach the negative electrode of the battery from the external circuit through the contact conductive solvent and battery current collector [10,11]. The ion transfer in the discharge process is opposite to that in the charge process. The specific positive and negative chemical reaction equations of LEP battery are as follows:

Positive reaction:

$$LiFePO_4 \rightleftarrows xLi^+ + xe^- + Li_{1-x}FePO_4 \tag{1}$$

Negative reaction:

$$6C + xLi^+ + xe^- \rightleftarrows Li_xC_6 \tag{2}$$

Total battery reaction:

$$LiFePO_4 + 6C \rightleftarrows Li_{1-x}\,FePO_4 + Li_xC_6 \tag{3}$$

The analysis of the reaction mechanism of the lithium ion battery shows that the heat generation mechanism of lithium ion power battery broadly comprises the following: (1) the heat generated by the electrochemical reaction inside the battery; (2) the ohmic resistance heat generated by the physical resistance of lithium ion when it is transferred in the positive material and electrolyte solution [12,13]; (3) the polarization internal resistance caused by the electrochemical reaction of the electrode; (4) the concentrated polarization internal resistance caused by the lithium ion transport process [14]. Thus, the heat generated mainly consists of four parts: chemical reaction heat Q_f, ohmic internal resistance heat Q_n, polarization heat Q_p and side reaction heat Q_s. Eq. (4) presents the total heat reaction equation.

$$Q_z = Q_f + Q_n + Q_p + Q_s \tag{4}$$

Q_z is the total heat of the lithium ion battery. The value of chemical reaction heat Q_f is negative when the battery is charged and positive when the battery is discharged. The equation of chemical reaction heat is as follows:

$$Q_f = nmQI(MF) \tag{5}$$

n is the number of all batteries; m is the mass of the battery electrode (unit: g); Q is the heat of electrochemical reaction of each battery (unit: J); I is the current generated during battery charging and discharging (unit: A); M is the molar mass of chemical elements in the battery (unit: g/mol); F is the Faraday constant.

Q_n is the heat generated by the current flowing through the conductive substance in the lithium ion battery. The calculation equation is as follows:

$$Q_n = I^2 R_n \tag{6}$$

In Eq. (6), I is the amount of current generated during the charging and discharging process (unit: A); R_n is the ohmic internal resistance generated during the charging and discharging process (unit: Ω). Ohmic heat is scalar, and the value of heat generated is positive.

Q_s is the unnecessary heat generated when the lithium ion battery is not working or is about to stop working. It is generally very small and can be ignored in the calculation.

Q_p is the heat generated by the current generated in the process of battery charging and discharging passing the polarization internal resistance. It is a scalar value, which is non-negative. The specific calculation equation is as follows:

$$Q_p = I^2 R_p \tag{7}$$

I is the current generated during the battery charging and discharging process (unit: A); R_p is the polarization internal resistance (unit: Ω).

The total calorific value of the lithium ion battery can be obtained from the above equations, as follows:

$$Q_z = Q_f + Q_n + Q_p + Q_s = \mathrm{nmQI(MF)} + \mathrm{I^2 R_t} \tag{8}$$

In (8), R_t is the sum of R_p and R_n in the battery.

When thermal runaway occurs in lithium ion batteries, the main processes of heat transfer to the outside world are thermal radiation, thermal convection and thermal conduction [15]. Heat transfer by thermal radiation is in the form of electromagnetic waves and can be calculated by the following equation:

$$Q_{\mathrm{rad}} = \varepsilon\sigma A(T_1^4 - T_a^4) \tag{9}$$

Q_{rad} is the total energy transferred by the thermal radiation process (unit: J); ε is the emissivity in the thermal radiation; σ is the Stefan-Boltzmann constant, with a fixed value of 5.67×10^{-8} W/(m^{2}K^4); T_1 is the surface temperature of the first battery in the lithium ion battery (unit: K); T_a is the external environment temperature of the lithium ion battery when working (unit: K); A is the surface area of the lithium ion battery (unit: m^2).

Thermal conduction is a hypothetical analysis of energy transfer during thermal runaway in a lithium ion battery. If the second battery of a lithium ion battery pack belongs to a single equilibrium object, when there is no thermal runaway taking place, the temperature of the first battery increases due to the thermal conduction effect. The specific calculation equation is as follows [16]:

$$Q_{cond} = \mathrm{C_p m}(T_2 - T_o) \tag{10}$$

Q_{cond} is the total energy transferred by the thermal conduction process (unit: J); C_p is the average specific heat capacity of the lithium ion battery (unit: $J \cdot g^{-1} \cdot k^{-1}$); m is the mass of the lithium ion battery (unit: g); T_2 is the surface temperature of the second battery of the lithium ion battery pack (unit: K); T_o is the initial temperature of the second battery, (unit: K).

Thermal convection is the heat transfer reaction generated by fluid flowing between the surface of the lithium ion battery and the environment around the battery when the thermal runaway occurs. The calculation equation is as follows [17]:

$$Q_{conv} = \mathrm{hA}(T_1 - T_a) \tag{11}$$

In Eq. (11), Q_{conv} is the total energy produced by the thermal convection process (unit: J); h is the convection coefficient in the thermal convection; A is the surface area of the lithium ion battery (unit: m^2).

2.2 Experimental Design of Water Mist Additive

Water mist additives can be divided into two categories according to their physical and chemical properties; namely, water-soluble salt additives and surfactants [18].

Water-soluble salt additives: These mainly contain alkaline metal salts, ammonium salts and other water-soluble chemicals. They can be further divided into two categories according to different action mechanisms. Inorganic salt additives, such as NaCl, $NaHCO_3$, KCl, and $KHCO_3$, can enhance the fire extinguishing effect of water mist (However, Na^+ and K^+ ions ionized from inorganic salts can combine with free radicals in the combustion chain; therefore, if additives containing inorganic salts are overused, the pipeline conveying the additives will be corroded) [19]. The other category is decomposition additives, which are easy to decompose. At a high temperature, these additives can decompose quickly to absorb heat, in order to achieve the purpose of cooling. Inorganic salts and decomposition additives can simultaneously enhance the effect of water mist to inhibit heat runaway. The most commonly used are $CO(NH_2)_2$ and $NH_4H_2PO_4$.

Surfactants: The most commonly used are quaternary ammonium surfactant (FC-4) and sodium dodecyl benzene sulfonate (SDBS), which can change the surface tension, particle size, adhesion and other physical properties of water mist droplets [20]. The cooling effect of water mist is improved by changing these physical properties, which is different from the action of water-soluble salt additives, which improve the combination with free radicals in the combustion chain. The ability of water mist to inhibit thermal runaway is limited. In the experiment reported in this paper, to control the thermal runaway reaction of a lithium ion battery, a water-soluble salt additive and a surfactant are selected for comparative study.

The experimental equipment used in this experiment is the HAIGINT sea view humidifying water mist system equipment, as shown in Fig. 2. In the experiment, the pressure is set to 2 MPa, the spray particle diameter of the water mist system is set at 100 μm, the spray cone angle of the system is set to 85°, and the flow rate of the water mist is 20 ml/s. It can automatically control the timing and the spraying time of the system; a large enough water tank is used to provide the water source for the experiment to explore the inhibition effect of different water mist additives. In order to ensure the accuracy of the experiment, the upper limit of the total amount of water mist additive is 300 ml, and the total application time is 20 s. The main purpose is to test the effect on a lithium ion battery with the thermal runaway phenomenon. When water mist additive is added to suppress thermal runaway, all external heat transfer and heating devices are removed, so as to avoid the influence of a temperature change in the external environment on the experimental results.

Figure 2: HAIGINT sea view humidifying water mist system equipment

Tab. 2 shows three kinds of additives selected for the experiment of inhibiting thermal runaway of a lithium ion battery by means of water mist, with the specific concentrations used.

Table 2: Selected additive schemes

Additives	Function	Category	Concentration (%)
NaCl	Enhance the fire extinguishing effect of water mist	Inorganic salts	10
SDBS	Improve the physical properties of water mist	Surfactants	0.2
$CO(NH_2)_2$	Decomposing to absorb heat and accelerate cooling	Decomposable class	7

3 Results and Discussion

3.1 Experimental Analysis of Heat Generating Rate of Li Ion Battery at Different Discharge Rates

In order to study the energy transfer of a lithium ion battery after thermal runaway, the corresponding state of charge (SOC) at different discharge rates is selected; that is, the heat generating rate of the battery with residual power (usually expressed as SOC percentage) from 10% to 100%. Tab. 3 shows the specific values.

Based on the data in Tab. 2, the function curves of the heat generating rate of the battery at different discharge rates are obtained, as shown in Fig. 3.

Fig. 3 reveals that the heat generating rate of lithium ion batteries changes with the discharge depth. The heat generating rate is the highest when the remaining power is 10%, and is lowest when the remaining charge is 100%. This is because lithium ion batteries mainly depend on the internal resistance to generate heat when their power is too low. Therefore, the internal resistance of lithium ion batteries is an important factor affecting internal heat generation. The height of the curve of the heat generating rate at 5C discharge rate is different from that at 10C discharge rate, but the trend of the curve is the same.

Table 3: Heat generation at different discharge rates

SOC (%)	Discharge rates	
	5C	10C
10	42964.48	155679.48
20	35489.63	127614.97
30	31016.52	112124.11
40	28994.62	103452.71
50	27178.34	99173.26
60	26895.74	98774.32
70	25748.41	98573.63
80	24657.63	98138.34
90	23987.49	97146.28
100	141.34	520.15

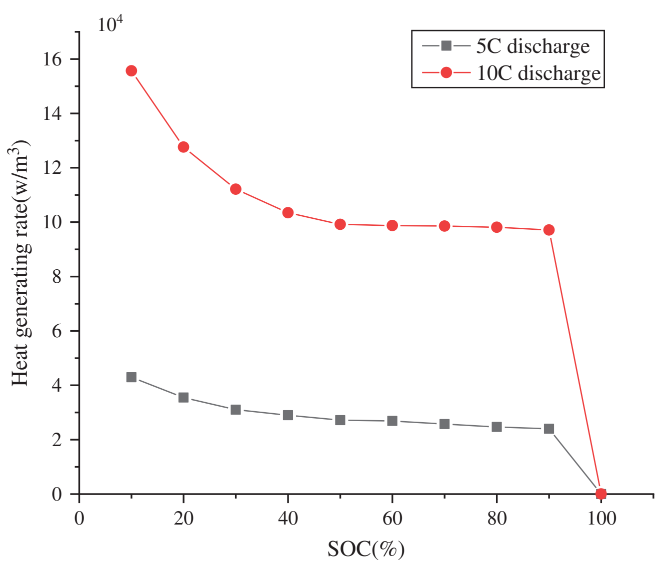

Figure 3: Variation of heat generation rate at different discharge rates

3.2 Analysis of Surface Temperature Change of Lithium Ion Battery at Different Charge and Discharge Rates

In order to explore the change of battery surface temperature at normal temperature (25°C) in the thermal runaway of lithium ion battery, three different charging rates are selected and the temperature acquisition instrument is used to measure the battery surface temperature, as shown in Fig. 4.

0.5C, 1C and 1.5C charging rates are selected in turn. Fig. 4 reveals that the surface temperature of the battery changes with the charging time; the temperature of the lithium ion battery increases with time. At the beginning of charging, the battery surface temperature rises the most, which is due to the larger internal resistance and lower internal heat dissipation of the battery at the beginning of charging. After the battery charging becomes stable, the resistance in the battery gradually decreases, and the heat generated by the internal resistance of the battery begins to decrease, gradually showing a trend of balance between heat dissipation and heat generation.

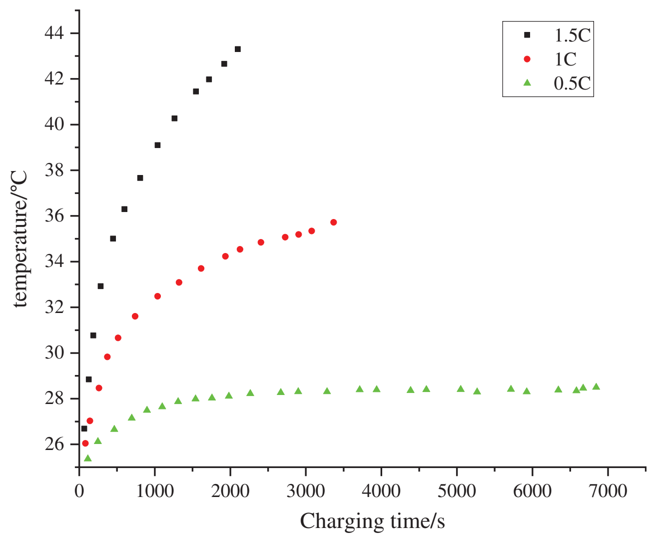

Figure 4: Surface temperature change of lithium ion battery under different charging rates

Moreover, the surface temperature of the battery varies with the charge rate. At 0.5C charging rate, the surface temperature of the battery rises slowly. As the charging time goes on, the surface temperature of the battery hardly increases. At 1C and 1.5C charging rates, the temperature curve increases with the charging time.

From the above analysis, it can be concluded that, in the process of constant current charging, the larger the battery charging rate, the higher the surface temperature of the battery, and the greater the corresponding power generation of the battery. However, a higher charging rate will lead to a high battery temperature, which may result in an explosion or failure of the battery. Therefore, it is necessary to control the charging current and the rate of rising temperature of the battery during the charging process.

Fig. 5 shows the change in the surface temperature of a lithium ion battery at 1C and 2C discharge rates. Image analysis reveals that the surface temperature of the battery increases with increasing discharge time at different discharge rates. The higher the discharge current is, the faster the surface temperature rises. In the later stage of battery discharge, the surface temperature of the battery shows a sharp upward trend. This is because the current increases continuously in the process of battery discharge, and the heating power of the battery also increases continuously, but the natural heat dissipation of the battery in the external environment does not change much. In the later stage of battery discharge, the internal resistance of the battery increases continuously, which causes a rapid increase in its heating power, resulting in a sharp increase in the surface temperature of the battery, as shown by a sharp increase in the temperature image in the later stages. At 1C and 2C discharge rates, it is obvious that the temperature rise at the 2C discharge rate is greater than that at the 1C discharge rate, which also shows that the discharge current has a great influence on the surface temperature and performance of the battery during the discharge process. If the discharge current continues to increase, the performance of the battery will deteriorate and the service life of the battery will be damaged.

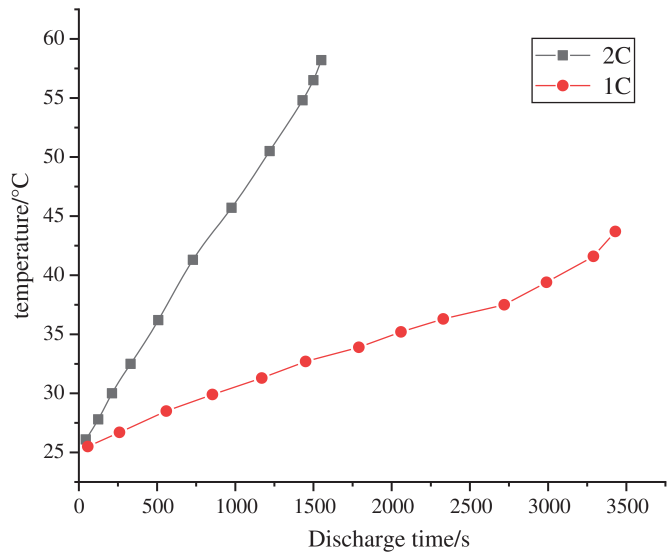

Figure 5: Surface temperature change of battery at different discharge rates

3.3 Experimental Analysis of Water Mist Suppression of Thermal Runaway in a Lithium Ion Battery

After the thermal runaway reaction of the battery occurs, it is necessary to turn on the water mist, and add the chosen additives into the water in turn. The surface temperature change and effect of each additive on the thermal runaway reaction of the battery are analyzed, as shown in Fig. 6.

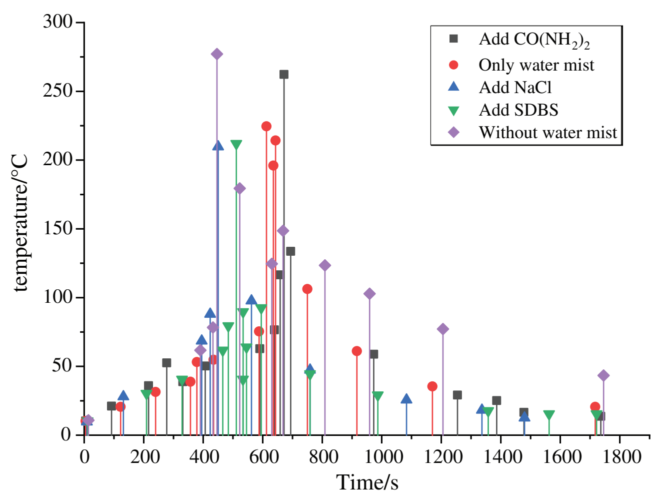

Figure 6: Temperature change of battery with different additives

Fig. 6 suggests that after the thermal runaway reaction, the maximum temperature of the battery with water mist is lower than without water mist, and the downward trend of the battery with water mist is greater than without water mist. It shows that adding water mist in the process of thermal runaway can inhibit the continuous reaction of chemical substances and reduce the exothermic reaction.

When NaCl, SDBS and $CO(NH_2)_2$ are added to the water mist to act on the battery, under the same conditions, the effect of NaCl is greater than that of SDBS and $CO(NH_2)_2$ to inhibit the thermal runaway reaction. Moreover, the temperature of the battery when NaCl is added shows the strongest downward trend and the fastest downward speed, which can effectively prevent the heat transmission process in the thermal runaway of the battery.

Finally, the optimum additive concentration of each common additive is obtained through comparative experiments with different concentrations of additives, as shown in Tab. 4.

Table 4: Optimum concentration of common additives

Additives	Optimum concentration %
SDBS	1.78
$CO(NH_2)_2$	4.8
NaCl	20

The change in the heat generating rate of the battery in a saturated and unsaturated state in discharging is explored. The experimental results show that the heat generating rate of the battery in 100% saturation state is much lower than in 10% saturation. The internal resistance of the battery is the main factor affecting the heat generating capacity of the battery when the battery power is too low. The surface temperature changes of the battery under different charge-discharge rates are explored. The results suggest that the higher the SOC, the worse the safety performance and the shorter the battery life. The inhibition effect of different water mist additives on the thermal runaway of battery is explored. The experimental results show that the temperature of the battery with 20% NaCl in the water mist additive leads to the greatest and fastest reduction in speed, which can effectively prevent the heat transmission process in the thermal runaway in the battery. The results of this experiment will be applied to lithium ion battery installations after technical improvement, which will effectively improve the service life of lithium ion batteries and inhibit the occurrence of the thermal runaway phenomenon.

4 Conclusion

The LEP form of the lithium ion battery is taken as the research object. The structure and reaction equation of the battery are briefly introduced. First, discharge rates of 5C and 10C are used to study the change in the heat generating rate of lithium ion batteries. Image analysis shows that the heat generating rate of the battery is lowest when the remaining power is 100%, and the highest when the remaining power is 10%. The surface temperature changes of the battery under different charge-discharge rates are explored. The results show that a higher charge and discharge rate leads to a higher surface temperature. Hence, the conclusion is that a lower discharge rate and charge/discharge current should be set to prolong the service life of the battery. Finally, water mist additives are used to inhibit the thermal runaway phenomenon of lithium ion batteries, and the inhibition effects of different water mist additives at different concentrations on thermal runaway are explored. It is found that water mist containing NaCl additive has the best inhibition effect on the thermal runaway of the lithium ion battery, and the inhibition effect is greatest when the concentration of the NaCl additive is 20%. This exploration mainly aims to solve the problem of the thermal runaway reaction in lithium ion batteries caused by poor heat dissipation performance. The results prove that using a water mist with 20% NaCl additive can inhibit the thermal runaway reaction in lithium ion batteries.

It is hoped that the performance of lithium ion batteries can be further improved through other research directions in future research, such as identifying factors that influence poor heat dissipation performance, so as to improve the service life of lithium ion batteries and reduce wastage of energy.

Funding Statement: The authors received no specific funding for this study.

Conflicts of Interest: The authors declare that they have no conflicts of interest to report regarding the present study.

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